LETTERS

# Southern Ocean deep-water carbon export enhanced by natural iron fertilization

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**The addition of iron to high-nutrient, low-chlorophyll regions induces phytoplankton blooms that take up carbon[1–3]. Carbon export from the surface layer and, in particular, the ability of the ocean and sediments to sequester carbon for many years remains, however, poorly quantified[3]. Here we report data from the CROZEX experiment[4] in the Southern Ocean, which was conducted to test the hypothesis that the observed north–south gradient in phytoplankton concentrations in the vicinity of the Crozet Islands is induced by natural iron fertilization that results in enhanced organic carbon flux to the deep ocean. We report annual particulate carbon fluxes out of the surface layer, at three kilometres below the ocean surface and to the ocean floor. We find that carbon fluxes from a highly productive, naturally iron-fertilized region of the sub-Antarctic Southern Ocean are two to three times larger than the carbon fluxes from an adjacent high-nutrient, low-chlorophyll area not fertilized by iron. Our findings support the hypothesis that increased iron supply to the glacial sub-Antarctic may have directly enhanced carbon export to the deep ocean[5]. The CROZEX sequestration efficiency[6] (the amount of carbon sequestered below the depth of winter mixing for a given iron supply) of 8,600 mol mol$^{-1}$ was 18 times greater than that of a phytoplankton bloom induced artificially by adding iron[7], but 77 times smaller than that of another bloom[8] initiated, like CROZEX, by a natural supply of iron. Large losses of purposefully added iron can explain the lower efficiency of the induced bloom[6]. The discrepancy between the blooms naturally supplied with iron may result in part from an underestimate of horizontal iron supply.**

In many open ocean regions there is low phytoplankton biomass despite there being a large macronutrient reservoir[3]. The Southern Ocean is the most biogeochemically significant of these high-nutrient, low-chlorophyll (HNLC) regions, owing to its large spatial extent and influence on global nutrient cycles[9]. Mesoscale iron enrichment experiments have demonstrated that iron addition modifies phytoplankton processes, enhancing diatom biomass[10,11] and increasing atmospheric carbon dioxide drawdown[1]. Observing bloom decline and quantifying the sequestration of photosynthetically fixed carbon resulting from iron addition has been achieved more rarely[7,12].

An alternative way to determine the role of iron in regulating the biological carbon pump in the Southern Ocean is to study regions of high phytoplankton biomass stimulated by natural iron inputs from shallow topography or islands. Recently KEOPS[8] (the Kerguelen ocean and plateau compared study) demonstrated enhanced carbon export to below 200 m in the naturally iron-fertilized bloom over the Kerguelen plateau. The Crozet Islands and Plateau (hereafter Crozet), located in the Polar Frontal Zone at the northern boundary of the Southern Ocean, is another region characterized by a marked annual phytoplankton bloom (Fig. 1). The sub-Antarctic Front of the generally eastward-flowing Antarctic Circumpolar Current turns north past Crozet (Fig. 1) and then east again when it encounters the Agulhas Return Current[13]. Thus, south of Crozet HNLC conditions prevail[4], whereas north of Crozet an annual bloom covering 120,000 km$^2$ (the size of Ireland and 50 times larger than the SOFeX (Southern Ocean iron experiment) bloom[2,11]) results from iron supplied from Crozet[14]. Iron enrichment over the light-limited winter period leads in spring to a strong north–south gradient in phytoplankton biomass (Fig. 1), productivity, community structure[15] and uptake of dissolved inorganic carbon[16] and nitrate[17], once stratification and increased solar irradiance reduce the mixed layer below the critical depth[18]. Weak circulation in the bloom region is such that water has a residence time there of ~60 days[4].

During austral summer 2004–2005, we conducted an extensive oceanographic research programme (CROZEX) around Crozet[4] to test the hypotheses that the north–south gradient in chlorophyll *a* is, first, induced by natural iron fertilization and, second, causes enhanced organic carbon flux into the deep ocean. To capture this flux, sediment traps were moored north (M10), east (M5) and south (M2, M6) of Crozet (Fig. 1). Short sediment cores were collected at M5, M6 and M10. Weak eastward flow past M2 and M6 and the absence of upstream blooms[13] characterized these HNLC 'control' (−Fe) sites south of the bloom. M10 was under the bloom (+Fe) and M5 was under the eastward extension of the bloom. East–southeast flow along the sub-Antarctic Front towards M5, the large spatial extent of the bloom combined with weak circulation within it and the predominance of *Eucampia antarctica* (a diatom that responded strongly to iron enrichment[19]) in the 3,000-m M10 and M5 traps (but

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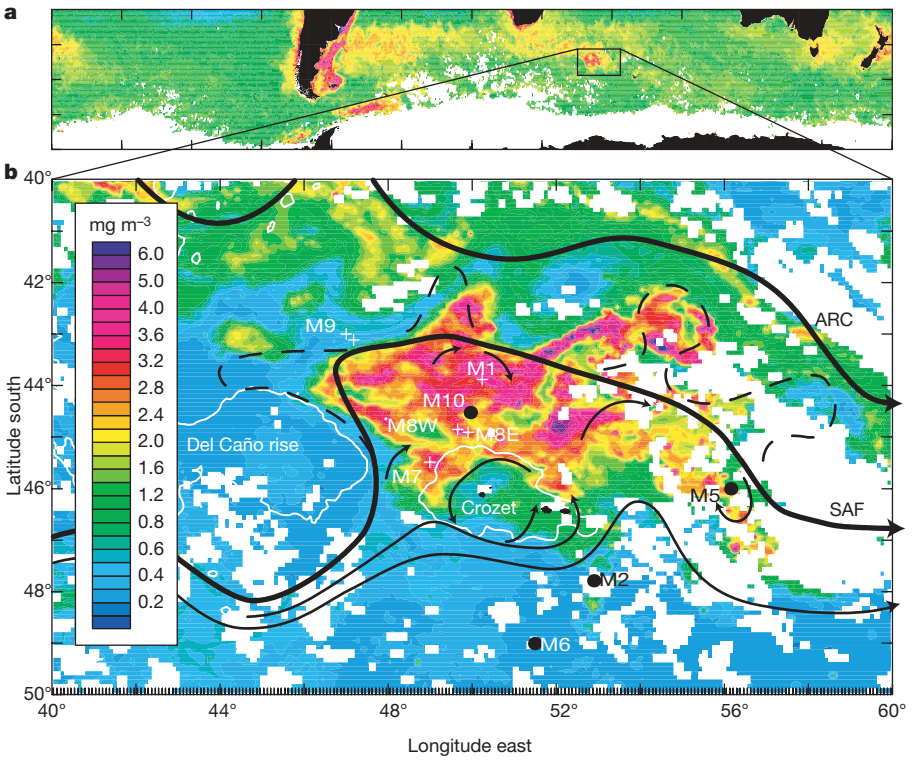


**Figure 1 | Chlorophyll *a* images of Crozet region.** **a**, Chlorophyll *a* in October for the whole of the Southern Ocean, showing location of Crozet. Colour indicates concentration as shown in **b**. **b**, Merged SeaWiFS/MODIS (sea-viewing, wide-field-of-view sensor/moderate-resolution imaging spectroradiometer) chlorophyll *a* image for the eight-day peak bloom period 23–30 October 2004. Solid and dashed lines show mean and eddy circulations, respectively[13], with the sub-Antarctic Front (SAF, the northern boundary of the Antarctic Circumpolar Current) and the Agulhas Return Current (ARC) shown bold. Main sampling (+) and coring (•) sites are labelled. Thin white lines are the 2,000-m depth contour, with the main Crozet Islands (Île de la Possession, Île de l'Est) seen at 46.5° S, 52° E.

its absence from the M2 and M6 traps) confirm that the M10 and M5 traps received export flux from the iron-enhanced bloom. KEOPS and CROZEX are compared in Supplementary Table 1.

It has been shown[14] that the dissolved iron (DFe; <0.2-μm fraction) originates from Crozet, with maximum estimated input to the bloom of 550 nmol $m^{-2}$ $d^{-1}$ comprising 390, 60 and 100 nmol $m^{-2}$ $d^{-1}$ for the horizontal, vertical and atmospheric fluxes, respectively. As the bloom occurs in deep (>2,000-m) water away from Crozet, horizontal flux dominates DFe supply, as expected. A range of 180–390 nmol $m^{-2}$ $d^{-1}$ (0.018–0.039 mmol $m^{-2}$ integrated over a winter period of 100 days) is estimated (Supplementary Information) for the enhancement in iron supply to the +Fe region relative to that to the −Fe region. These are probably underestimates, as additional sources of iron such as the dissolution of small lithogenic particles[20] will increase iron supply.

Significant differences were observed in the magnitude, timing, duration and community structure of plankton blooms between the +Fe and −Fe regions. In the −Fe region, chlorophyll *a* peaked at 0.6 mg $m^{-3}$ in early December (Fig. 2a), when HNLC conditions (nitrate concentration, ~24 μmol $kg^{-1}$; silicate concentration, ~16 μmol $kg^{-1}$) prevailed[4]. In the +Fe region, chlorophyll *a* peaked at over 3 mg $m^{-3}$ in October (locally >6 mg $m^{-3}$; Fig. 1) and was elevated (>1 mg $m^{-3}$) for 72 days[18] (Supplementary Table 2). Although fertilized by macronutrients from the −Fe region and by winter upwelling in the Polar Frontal Zone, silicate was already becoming limited (<2 μmol $kg^{-1}$; nitrate, 16 μmol $kg^{-1}$) when first sampled in November[4], indicating a ratio of silicate drawdown to nitrate drawdown of about 2:1, consistent with lower iron stress than in the −Fe region[19]. Low ambient silicate concentrations, common over much of the sub-Antarctic Southern Ocean[11], predisposed a shift in phytoplankton community structure from diatoms to *Phaeocystis*[15]. The bloom peaked <10 days after exceeding

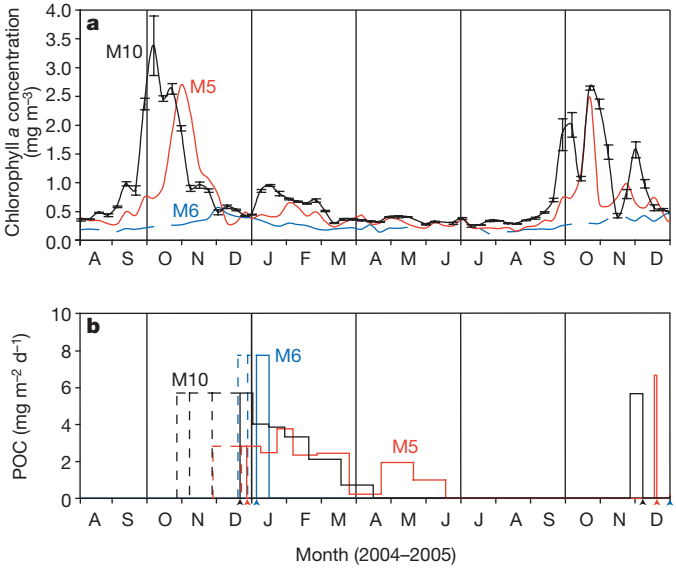


**Figure 2 | Time series of chlorophyll *a* and particulate organic carbon (POC).** **a**, Chlorophyll *a*, obtained for each eight-day merged SeaWiFS/MODIS image by averaging all non-cloud pixels in a circle of radius 45 km about each mooring site. Error bars (for M10 only) give the standard deviation of the mean of these pixels. **b**, POC (solid line) obtained from deep sediment traps at each site (Methods). Arrowheads mark mooring deployment and recovery events. The traps could not be deployed until after export from the 2004–2005 bloom had begun, so the export rate into the first cup has been extrapolated (Supplementary Information) using a range of sinking rates after the peak of chlorophyll *a* to give minimum, mean and maximum (dashed) seasonal integrals of total export.

$1\,mg\,m^{-3}$ but remained $>1\,mg\,m^{-3}$ for another month, potentially sustained by *Phaeocystis* using regenerated iron and nitrogen[21], as +Fe nitrate values remained $\sim16\,\mu mol\,kg^{-1}$ throughout December and January.

An important difference from purposeful iron enrichment experiments is that iron concentrations accumulate in the +Fe region during winter. Removal of light limitation in spring[18], not iron addition, determines bloom onset. Consequently, net growth rates in the bloom phase ($0.05\,d^{-1}$; Fig. 2a) are probably light limited and 2–18 times lower than those for artificial experiments ($0.10–0.90\,d^{-1}$)[3]. Weak circulation in the +Fe region ensures that neither macronutrients nor iron can be resupplied to the extensive bloom area during the bloom development period. A possible exception was close to the islands, where a small-area bloom in January[18] may have been fuelled by resupply of iron and silicate.

The flux of organic carbon from the surface ocean to the ocean interior has been calculated using $^{234}$Th at 100 m (ref. 22; Table 1). Following the chlorophyll peak (Fig. 2a) in each region (+Fe, −Fe), mean daily rates of carbon export were similar ($16\,mmol\,m^2\,d^{-1}$; Supplementary Table 2). Thus, any difference in seasonally integrated export between the two regions depends on the duration of the export events. We estimated export duration by closing the silicate budget, dividing the near-surface silicate drawdown (corrected for biogenic silica production) by the opal export rate estimated from $^{234}$Th deficits and $^{234}$Th/opal ratios. This approach yielded export durations (61 and 17 days in the +Fe and −Fe regions, respectively) consistent with the observed satellite-derived chlorophyll time series (Fig. 2, Supplementary Table 2). The resulting seasonally integrated carbon export in the +Fe region ($960\,mmol\,m^{-2}$) was three times greater (Table 1) than export in the −Fe region ($290\,mmol\,m^{-2}$), consistent with the independently diagnosed increase in new production[17].

Fluxes of particulate organic carbon (POC) to 3,000 m differed remarkably in duration and composition. In the +Fe region (Fig. 2b), POC flux peaked at or before trap deployment in late December, decreasing to near zero over several months. In the −Fe region, POC export was confined to an unusually short but substantial event (Fig. 2b) observed at both M2 and M6 and at two depths (for sinking rates and export flux ranges, see Supplementary Information). Substantial silicate drawdown between November and January (Supplementary Table 2) reduced surface silicate to $<2.0\,\mu mol\,kg^{-1}$ at M2 and M6, suggesting that iron limitation in the −Fe region resulted in heavily silicified diatoms[23] that sank rapidly in January. Despite this event, the longer duration of POC flux over the 2004–2005 summer season in the +Fe region resulted in three times greater seasonal export in the +Fe region than the −Fe region (Table 1). POC flux at 3,000 m was 3% of that at 100 m in the +Fe region and 4% of that at 100 m in the −Fe region (Table 1), indicating that remineralization rates were marginally enhanced by iron availability.

**Table 1 | Seasonally integrated carbon fluxes at naturally iron fertilized and HNLC sites and the sequestration efficiency, C/Fe**

| | Carbon ($mmol\,m^{-2}\,y^{-1}$) | | C/Fe‖ ($mol\,mol^{-1}$) |
|---|---|---|---|
| | +Fe (fertilized) | −Fe (HNLC) | |
| $^{234}$Th via Si* at 100 m | 960 | 290 | 17,190 |
| Range | 626–1,252 | 166–415 | 5,420–60,360 |
| Deep flux† at 3,000 m | 25.0 | 7.1 | — |
| Best estimate‡ | 28.9 | 11.6 | 440 |
| Range‡ | 25.0–34.2 | 7.1–17.4 | 195–1,506 |
| Core top§ | 9.3 ± 0.5 | 4.5 ± 0.4 | 123 |
| Interpolated flux at 150 m¶ | 642 | 194 | 11,487 |
| Interpolated flux at 200 m¶ | 483 | 146 | 8,641 |

* Summarized from Supplementary Table 2.
† From Fig. 2.
‡ Summarized from Supplementary Information.
§ Summarized from Supplementary Table 3.
‖ Calculated from the differences between +Fe and −Fe carbon fluxes divided by winter-period iron supply ($0.018–0.039\,mmol\,m^{-2}$).
¶ Calculated from 100-m flux ($F$) values using a Martin curve $F(z) = F(100\,m) \times (z/100)^b$, where $b = -0.99$ to fit the 3,000-m carbon flux values.

The organic carbon content of the core-top (surface-mixed-layer) sediments sampled several times during separate corer deployments was significantly higher in the +Fe region than the –Fe region (Supplementary Table 3). Significant sediment focusing and winnowing occurs in this region and thus data are expressed as $^{230}Th_{xs}$-corrected, preserved fluxes (equivalent to the preserved vertical rain rate at the sea floor). A twofold increase in the $^{230}Th_{xs}$-corrected, preserved, core-top, organic carbon accumulation was observed in the +Fe region relative to the –Fe region (Table 1). This is consistent with published data from a suite of export production proxies that imply enhanced phytoplankton growth, export and burial throughout the Holocene epoch at this site[24].

Our analyses thus indicate that shallow, seasonally integrated export, annually integrated deep-water POC flux and core-top organic carbon accumulation were all enhanced two- to threefold as a result of the iron-fertilized bloom (Table 1). Our results support Martin's hypothesis[5] that relief of iron deficiency enhances carbon sequestration into the deep ocean (here >3,000 m) and sediment. Results from CROZEX thus support increased atmospheric iron deposition[1] as a mechanism for the inferred increase in organic carbon flux in the sub-Antarctic during the Last Glacial Maximum[24,25].

The ratio of carbon exported below some depth to iron added at the surface, (C/Fe) is termed the export efficiency or (if below the depth of winter mixing) sequestration efficiency[6,8]. Our $^{234}$Th-derived estimates of the seasonal enhanced (+Fe minus −Fe) POC flux at 100 m ($670\,mmol\,m^{-2}$) and additional iron supply ($0.039\,mmol\,m^{-2}$) lead to a C/Fe ratio (at 100 m) of 17,200 $mol\,mol^{-1}$ (range, 5,400–60,400; Table 1). This value for the shallow export efficiency from CROZEX was somewhat higher than comparable values from iron-addition experiments (6,600 for SOFeX[12], 1,200 for SERIES (the sub-Arctic ecosystem response to iron enrichment study)[7]). Interpolating with a Martin curve[26] to a winter mixed-layer depth of 150–200 m (ref. 18), we further calculated a seasonal C/Fe sequestration efficiency of 11,500–8,600 (Table 1), compared with previous estimates of 500–3,300 (refs 7, 12) and the KEOPS[8] seasonal estimate of 668,000. Given the different methods used to estimate both additional iron supply and carbon export between studies[3,7,8,12], the reasons for the wide range of export efficiencies are unclear. However, we note that the KEOPS result depends on a combination of an eightfold-lower estimate for seasonal iron supply and a tenfold-higher estimate for carbon export (Supplementary Table 1). It is possible that iron supply was higher to the KEOPS bloom before the late-summer observation period on which the seasonal iron supply was based, owing either to enhanced vertical supply before surface-water stratification in spring, coupled with luxury iron uptake[27] (that is, more than is absolutely necessary), or to horizontal input of lithogenic material from nearby islands[28,29].

The results from CROZEX indicate that natural iron fertilization enhanced new production[17] and near-surface export at 100 m two- to threefold (Table 1). Moreover, we present evidence that carbon fluxes at 3,000 m and the sediment were similarly two to three times higher beneath the natural fertilized region than for a nearby HNLC region with similar end-of-winter macronutrient concentrations. Carbon sequestered past 200 m was only 50% of that exported past 100 m. Although the CROZEX estimate of carbon sequestration for a given iron supply was 20 times that of SERIES[7], it still falls 15–50 times short of some geo-engineering estimates[6], with significant implications for proposals to mitigate the effects of climate change through purposeful addition of iron to the ocean.

## METHODS SUMMARY

Chlorophyll was determined using remote-sensing techniques referenced to *in situ* data. Iron concentrations in the bloom were estimated using a simple model including horizontal and vertical advection and atmospheric deposition. Organic carbon and opal export rates were determined using $^{234}$Th deficits

and $^{234}$Th/opal and organic carbon ratios from large particles. Biogenic silica was determined by spectrophotometric analysis of silicate levels in digested filtered samples. Sediment traps were McClane traps. Core data were derived from analysis of multiple gravity and Megacorer-derived samples.

**Full Methods** and any associated references are available in the online version of the paper at www.nature.com/nature.

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**Supplementary Information** is linked to the online version of the paper at www.nature.com/nature.

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**Author Information** Data are held at the British Oceanographic Data Centre (http://www.bodc.ac.uk). Reprints and permissions information is available at www.nature.com/reprints. Correspondence and requests for materials should be addressed to R.J.S. (rics@noc.soton.ac.uk).

## METHODS

**Chlorophyll *a*.** Chlorophyll *a* (Figs 1 and 2) was determined from NASA's merged SeaWiFS/MODIS products, adjusted to match *in situ* data[18].

**Iron concentrations in the bloom region[14].** Iron concentrations were estimated by considering lateral advection of DFe from the islands into the surrounding water, vertical mixing of iron from beneath the seasonal thermocline and atmospheric deposition. Total dissolved iron concentrations (DFe $<$0.2-μm fraction) were determined using flow-injection catalytic spectrophotometric detection. Horizontal iron flux was estimated from samples of DFe collected along a series of stations extending seawards from the northern coast of Île de la Possession and by using the terrestrially derived, short-lived radium isotopes $^{223}$Ra and $^{224}$Ra to estimate horizontal mixing coefficients at the same stations[30]. Horizontal gradients in these species were combined with estimates of the plateau circumference to estimate total DFe release from the plateau over the 100-day winter period when the mixed-layer depth is such that the surface ocean is in contact with plateau sediments. Vertical iron flux was diagnosed from analysis of $^{228}$Ra and DFe profiles. Finally, the estimated atmospheric (wet and dry) iron flux was based on calcium and silicon concentrations in aerosols and DFe measurements in rain samples by ICP-OES (inductively coupled plasma-optical emission spectrometer). The flux of DFe to surface waters was extrapolated to, and integrated over, the winter period. Values are consistent with atmospheric dust transport models.

**Organic carbon and opal export.** Shallow rates were estimated by multiplying the observed 0–100-m deficit of the short-lived natural radioisotope $^{234}$Th by the $^{234}$Th/POC or $^{234}$Th/Opal ratio in large-volume samples of large particulate material ($>$53 μm) collected using a Stand-Alone Pumping System deployed approximately 20 m below the mixed layer[22].

**Biogenic silica.** Measurements of biogenic silica were made on 1-litre seawater samples filtered onto 0.4-μm polycarbonate filters, stored at $-20\,^{\circ}$C, digested in 0.2 mol sodium hydroxide and neutralized with 0.1 mol hydrochloric acid[31,32] and analysed using a Skalar Sanplus autoanalyser. Opal accumulation was estimated by integrating values in the upper 100 m.

**Sediment traps.** Traps were McLane 21-cup time-series arrays deployed on bottom-tethered moorings. Sampling bottles were filled with buffered preservative solution according to Joint Global Ocean Flux Study protocols. Upon recovery, samples were filtered through a 1-mm Nitex mesh. Swimmers were carefully removed from the $<$1-mm fraction. The $>$1-mm fraction was comprised exclusively of large swimmers. Some cups were contaminated by fish (*Notolepis coatsi*) feeding on the sinking material. All fish debris was picked out by hand. All chemical analysis was performed on the $<$1-mm fraction after it had been freeze-dried and homogenized. Following acidification with hydrochloric acid, particulate organic carbon and nitrogen were measured using a Carlo Erba NA 1500 elemental analyser following standardization with acetanilide.

**Core-top accumulation rates.** A Megacorer was used to obtain sediment cores with an undisturbed sediment–water interface and gravity core deployments were used to sample deeper sediments. Samples from the surface mixed layer (0–10 cm below the surface) were dried, ground and subjected to the same methodology as described for the sediment trap material for particulate organic carbon and nitrogen at the National Oceanography Centre, Southampton, and NERC Isotope Geosciences Laboratory. U-series isotopes were determined by isotope dilution multi-collector ICP-MS (inductively coupled plasma mass spectrometry) at the Department of Earth Sciences, University of Oxford. $^{230}$Th-normalized, preserved organic carbon fluxes were estimated from the sediment composition and the $^{230}$Th-normalized sediment accumulation rate (measured $^{238}$U/$^{232}$Th activity ratio of detrital end-member is 0.9). These data supplement published data[24] also tabulated for comparison.

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